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Phonons in *fcc* binary alloys

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Abstract : Born-Mayer potential has been modified to account for the unpaired (three body) forces among the common nearest neighbours of the ordered binary *fcc* alloys i.e. $Ni_{.3}Fe_{.7}$, $Ni_{.5}Fe_{.5}$ and $Ni_{.75}Fe_{.25}$. The three body potential is added to the two body form of Morse to formalise the total interaction potential. Measured inverse ionic compressibility, cohesive energy, lattice constant and one measured phonon frequency are used to evaluate the defining parameters of the potential. The potential seeks to bring about the binding among 140 and 132 atoms though pair wise (two body) and non-pair wise (three body) forces respectively. The phonon-dispersion relations obtained by solving the secular equation are compared with the experimental findings on the aforesaid alloys.

Keywords : Morse potential, phonon-dispersion, binary *fcc* alloys, dynamical matrix, paired and unpaired interactions.

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1. Introduction

The model potential studies (Batterman 1957, Moss 1969, Clapp and Moss 1968, Schwartz and Cohen 1964) have usually accounted for the two body forces while analysing the Cu-Au system of binary alloys for short range order parameter. Some of the studies (Khwaja *et al* 1977, Khwaja *et al* 1978, Bieber and Gautier 1984) have focussed the importance of many body forces in binary alloys. The various studies (Winter *et al* 1986, Thakur *et al* 1987, Kumar *et al* 1982) pertaining to the electronic structure of alloys suggest the importance of unpaired forces. The study of phonon-dispersion has been of central importance because it is very much sensitive to the changes in the electronic behaviour of alloys and its full description needs all the effective interactions to be considered explicitly in the theory. Weber (1982) has emphasised the importance of pairwise forces while explaining the phonon dispersion in alloys. Garg and Gupta (1985), Garg *et al* (1985) and Gupta and Gupta (1988) have employed de-Launay angular forces for explaining the role of unpaired forces in alloys. These angular forces however bear one to one identity with the first derivative of central pairwise potential. Further these forces fall short of the stability requirements and make use of such input data which are derived essentially from the experimental findings on phonon dispersion. In the wake of these inadequacies we have developed a model which

blends the pairwise and the non-pairwise forces in a more logical and consistent manner. The model, beside satisfying the equilibrium condition, employs a minimum number of input data.

The present communication is organised in four sections. Section 2 deals with the theory of the model. Section 3 is devoted to the computation and results. The conclusions are given in the Section 4.

2. Theory

2.1. The two body potential :

We have represented the two body generalised potential in the form given by Morse (1929) because once defined this potential is capable of explaining the pairwise coupling among any number of neighbours without asking for more input data. Further mathematical operations happen to be simple and exact with the exponential form of the potential. The potential may be expressed as,

$$\phi^{(2)} = \frac{D}{2(m-1)} \sum_j [\beta^m \{\exp(-m\alpha r_j)\} - m\beta \{\exp(-\alpha r_j)\}] \tag{1}$$

where r_j is the position coordinate of the j-th atom, D and α are the depth and strength parameters of the potential respectively. The parameter β of the potential embodies both α and r_o i.e.

$$\beta = \exp(\alpha r_o) \tag{2}$$

The exponent m describes the depth of the potential and incorporates indirectly such modifications of the two body potential as demanded by the exchange correlation effects due to electrons. Present study finds the appropriate value for m (= 1.1), which presents the best agreement with the experimental data on phonon dispersion.

2.2. The three body potential :

The Born-Mayer (1932) potential assumes a finite value at zero separation (r) of the atoms. To remove this deficiency one may consider its inverse dependence on separation (r), the modified form may be written as,

$$\phi^3 = \frac{Ae^2}{r} \exp(-Kr^2) \tag{3}$$

where A and K are the defining parameters of the potential. Following Sarkar and Sengupta (1969), one may express the spatial three body forces operating among the common nearest neighbours (lk, $l'k'$, $l''k''$) of the *fcc* structure i.e.

$$\phi^3 = \sum_{\substack{lk \\ l'k' \\ l''k''}} \gamma [r_1 (l'k', lk)\, r_2 (l''k'', lk)]\, r_o^{-2} \tag{4}$$

Here (l', k') and (l'', k'') the indices of the atoms which are common nearest neighbours of

the atom having index (l, k). While accomplishing the summation in eq. (4) we have considered all those trio of the atoms which conform with the following constriant,

$$r(lk) - r(l'k') = r(lk) - r(l''k'') \tag{5}$$

The atom (l, k) is centered at first neighbour and possible other first, second, third and fourth neighbours satisfying the eq. (5) are identified. In eq. (4), r_1 and r_2 are the equal separations among the atoms $l'k'$, $l''k''$ from the atom lk. Further r_0 is the equilibrium separation and γ is the second derivative of the modified Born-Mayer (eq. 3) potential.

The trio of atoms satisfying the eq. (5) fall under the following four groups.

(a) Forty eight atoms from among those located at origin and at first neighbouring sites.

(b) Twenty four atoms from among those seated at origin, at first and second neighbouring sites.

(c) Forty eight atoms from among those positioned at origin, first neighbouring and third neighbouring sites.

(d) Twelve atoms from among those situated at origin, first neighbouring and fourth neighbouring sites.

2.3. *The dynamical matrix—two body contribution :*

Two body contribution to the dynamical matrix may be derived in the usual manner i.e.

$$D_{ii}^{(2)}(\vec{q}) = 4(\beta_1 + 2\alpha_1) - 2(\beta_1 + \alpha_1) C_i (C_j + C_k) - 4\alpha_1 C_j C_k + 4\beta_2 S_i^2 + 4\alpha_2 (S_j^2 + S_k^2) \tag{6}$$

$$D_{ij}^{(2)}(\vec{q}) = 2(\beta_1 - \alpha_1) S_i S_j \tag{7}$$

where $C_i = \cos(\eta\, a\, qi / 2)$ and $S_i = \sin(\eta\, a\, qi / 2)$, q_i is the i-th component of the phonon wave vector $\vec{q}$, α_1 and α_2 are the first and β_1 and β_2 are the second derivatives of the two body potential.

2.4. *The three body contribution to dynamical matrix*

We find following relations for the diagonal $D_{ii}^{(3)}(\vec{q})$ and the off diagonal $D_{ij}^{(3)}(\vec{q})$ elements of the three body dynamical matrix i.e.

$$D_{i}^{(3)}(\vec{q}) = k_1 \gamma (1 - C_j C_k) \tag{8}$$

$$D_{ij}^{(3)}(\vec{q}) = k_2 \gamma\, S_i S_j \tag{9}$$

where γ as said earlier is the second derivative of potential.

2.5. *The dispersion relations :*

The phonon frequencies are obtained by solving the usual secular equation i.e.

$$D_{ij}(\vec{q}) - 4\pi^2 \upsilon^2 M I = 0 \tag{10}$$

where M is the mass of the atom and I is the unit matrix of 3×3 order.

Eq. (10) yields the following dispersion relations along the three major symmetry directions.

along [100]

$$4\pi^2 M \upsilon_L^2 = [D_{ii}(\vec{q})]_{(100)} \tag{11}$$

$$4\pi^2 M \upsilon_I^2 = [D_{ij}(\vec{q})]_{(100)} \tag{12}$$

along [110]

$$4\pi^2 M \upsilon_L^2 = [D_{ii}(\vec{q}) + D_{ij}(\vec{q})]_{(110)} \tag{13}$$

$$4\pi^2 M \upsilon_I^2 = [D_{ii}(\vec{q}) - D_{ij}(\vec{q})]_{(110)} \tag{14}$$

$$4\pi^2 M \upsilon_{T_2}^2 = [D_{RR}(\vec{q})]_{(110)} \tag{15}$$

along [111]

$$4\pi^2 M \upsilon_L^2 = [D_{ii}(\vec{q}) + 2D_{ij}(\vec{q})]_{(111)} \tag{16}$$

$$4\pi^2 M \upsilon_T^2 = [D_{ii}(\vec{q}) - D_{ij}(\vec{q})]_{(111)} \tag{17}$$

Eq. (11) to eq. (17) are fed with the appropriate expression of dynamical $D_{ij}^{(2)}(q)$ and $D_{ij}^{(3)}(q)$ in order to get respectively the two body and three body contribution towards the phonon dispersion.

3. Computations and results

In order to evaluate the defining parameter α, D and r_o of the generalised Morse potential (eq. 1), first the potential is made to satisfy the condition of lattice stability i.e.

$$\frac{d\phi^{(2)}}{d r_1} = 0 \tag{18}$$

Next the relation for inverse compressibility, a function of $\phi^{(2)}$ is fed with such values of β which reproduce the observed magnitude of the inverse compressibility. Eq. (18) actually gives rise to β in terms of α and r_1 and the parameter D identifies with the observed cohesive energy. Vegard's law for the given concentration of Ni and Fe is exploited to determine the lattice constant, inverse compressibility and cohesive energy of the alloys of present interest. For the detailed procedure of parameter evaluation, one may refer to Grifalco and Weizer (1959).

For determining the parameters A and k of the modified Born-Mayer potential (eq. 3) again it is subjected to lattice equilibrium i.e.

$$\frac{d\phi^{(3)}}{dr} = 0 \tag{19}$$

and then an exact fit to the measured phonon frequency at the wave vector (1, 0, 0) leads to the evaluation of these parameters (A, K). The input data and the computed parameters for

the said *fcc* alloys i.e. $Ni_{.3}Fe_{.7}$, $Ni_{.5}Fe_{.5}$ and $Ni_{.75}Fe_{.25}$ are shown in Table 1. Table 2 enlists

Table 1. Input data and computed parameters for m = 1.1

Alloy	Semilattice constant a [Å]	Bulk modulus k [10^{12}dyn/cm^2]	Cohesive energy ϕ [10^{-12}ergs]	D [10^{-12}ergs]	α (Å)$^{-1}$	r_o (Å)	A 10^{10}ergs	K (Å)$^{-2}$
$Ni_{.3}Fe_{.7}$	1.793	1.326	6.924	0.777	2.015	2.675	55.581	–0.404
$Ni_{.5}Fe_{.5}$	1.793	1.266	6.976	0.765	1.968	2.687	0.108	–0.404
$Ni_{.75}Fe_{.25}$	1.776	1.478	7.040	0.812	2.091	2.638	0.007	–0.411

Table 2. The computed force constants at $m = 1.1$ [10^4dyn/cm]

Alloy	α_1	α_2	β_1	β_2	γ
$Ni_{.3}Fe_{.7}$	–0.129	0.064	3.031	–0.234	0.014
$Ni_{.5}Fe_{.5}$	–0.133	0.063	2.930	–0.217	0.029
$Ni_{.5}Fe_{.25}$	–0.129	0.067	3.295	–0.264	0.014

the computed force constants (α_1, α_2, β_1,β_2) and γ for these alloys. Figures 1, 2 and 3depict the computed phonon-dispersion for the ordered binary *fcc* alloys i.e. Ni_3Fe_7,

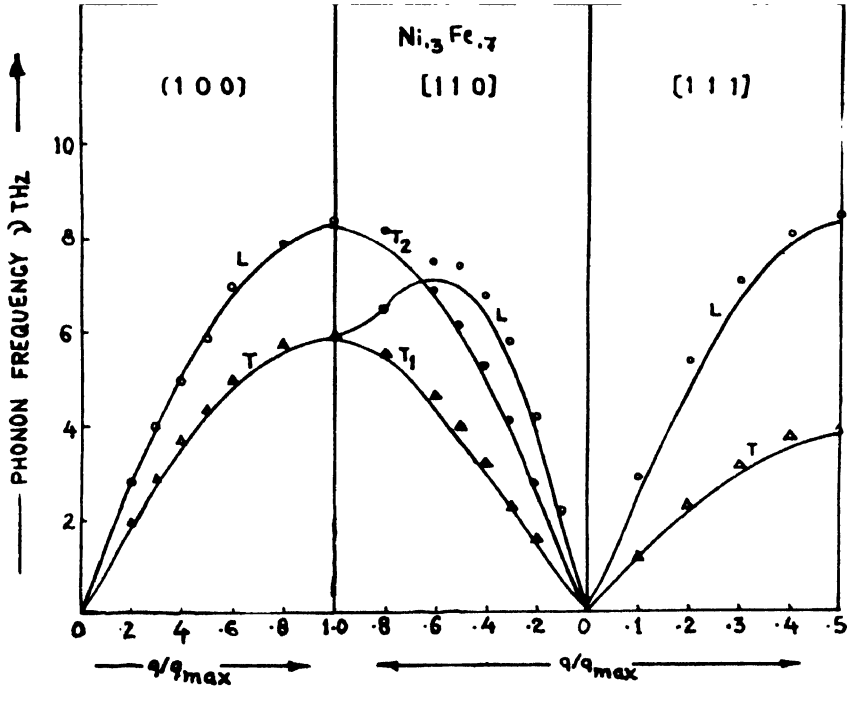

Figure 1. Phonon dispersion curve for Ni_3Fe_7.

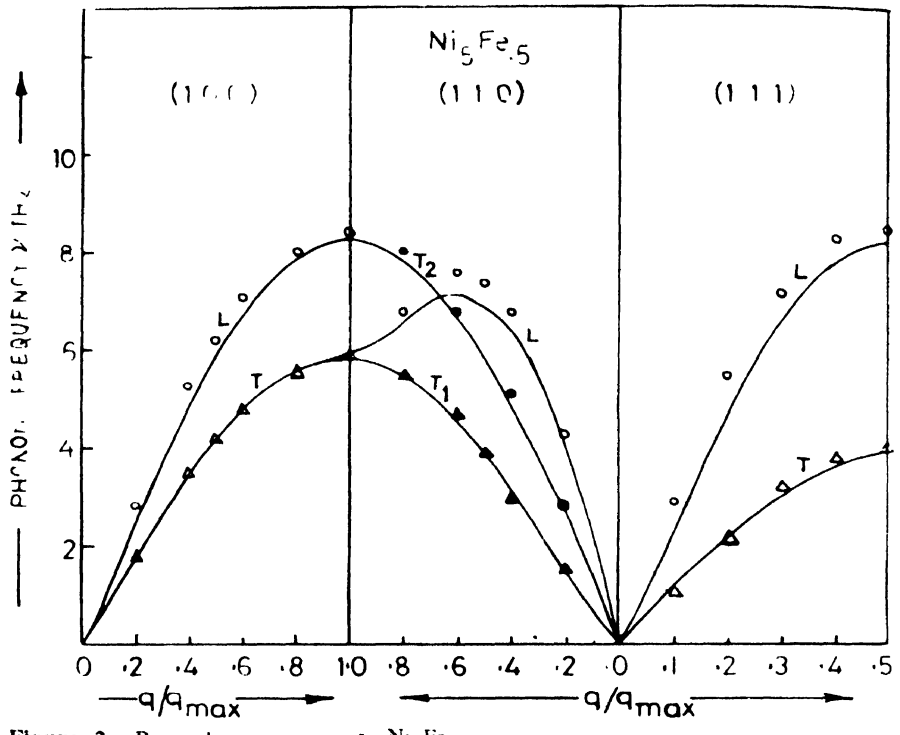

Figure 2. Ponon dispersion curve for Ni_5Fe_5.

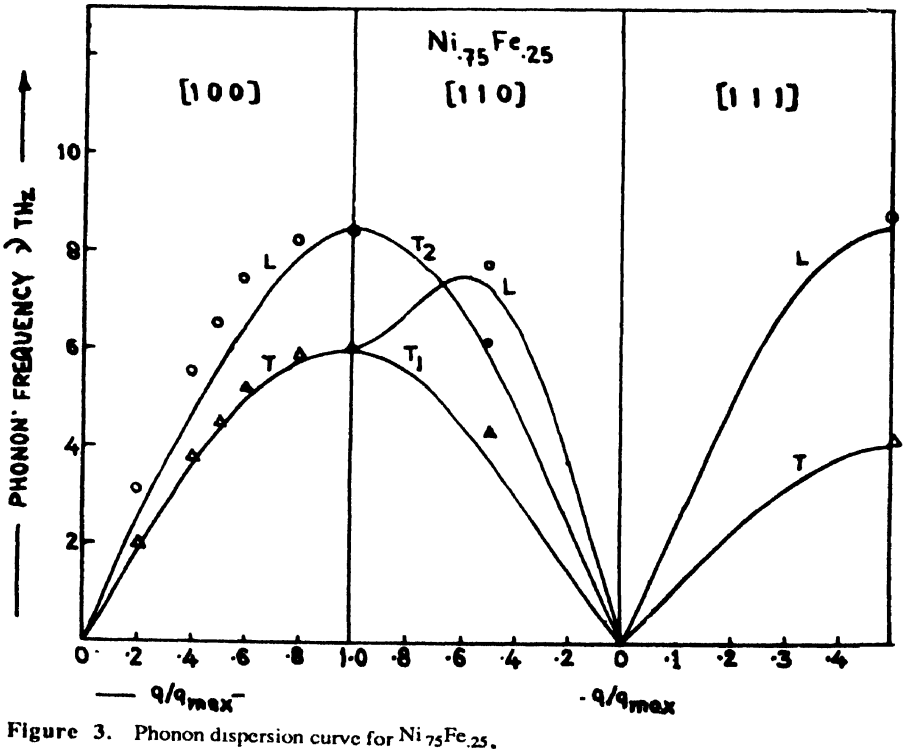

Figure 3. Phonon dispersion curve for $Ni_{75}Fe_{.25}$.

Ni_5Fe_5 and $Ni_{75}Fe_{25}$ respectively. Marking (O, Δ, ●,▲) indicate the measured data on phonon dispersion for Ni_3Fe_7, Ni_5Fe_5 and $Ni_{75}Fe_{75}$ reported by Hollman and Brockhouse (1969).

4. Conclusion

Perusal of Figures 1-3 reveals that our results on these alloys turn out to be satisfactorily close to the recent experimental findings of Hollman and Brockhouse (1969).

It is therefore evident that the present scheme of the modified Born-Mayer potential for three body interactions, when combined with generalised Morse potential for two body interactions explain fairly well the unpaired three body and the paired two body interactions in ordered binary *fcc* alloys of Nickel-Iron concentrations.

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